



July 19, 1990

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Hon. Commissioner of Patents & Trademarks
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RE: U.S. Patent Application of Richard R. Hertzog et al.
Title: DECOMPOSITION OF CUMENE HYDROPEROXIDE
Serial No. 297,333; Filed January 17, 1989
PD File No.: 30-2004 (4690)

Sir:

91-0763

Enclosed in triplicate is a Brief on Appeal in
connection with the above-referenced application.

The fee for filing this Brief on Appeal is
\$140.00. Please charge all fees to Account No. 01-1125; a
triplicate copy of this letter is also enclosed.

Respectfully,

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Enclosures

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126



PATENT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

Art Unit: 126

Examiner: J. Reamer

P.D. File No.: 30-2004 (4690)

Inventors: RICHARD R. HERTZOG ET AL.

Serial No.: 297,333

Filed: January 17, 1989

For: DECOMPOSITION OF CUMENE HYDROPEROXIDE

Petersburg, Virginia 23804
July 19, 1990

Commissioner of Patents
Washington, DC 20231

Sir:

BRIEF ON APPEAL

91-0763

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This is a brief on appeal from the Examiner's Action dated February 15, 1990.

(1) Status of Claims. No claims stand allowed. Claims 1-8 stand rejected. The claims on appeal, claims 1-8, are set forth in Appendix A, attached hereto.

(2) Status of Amendments. There is no amendment filed subsequent to the final rejection.

(3) Summary of the Invention. Phenol is manufactured via air oxidation of cumene to cumene hydroperoxide (CHP), followed by acid-catalyzed cleavage of CHP to phenol and acetone. CHP decomposition is a very exothermic reaction which is normally carried out on a commercial scale in continuous stirred or back-mixed reactors. In such reactors only a small fraction of CHP remains at any given time and the reaction medium consists essentially of the products of decomposition of CHP, i.e., phenol and acetone, plus any solvent (e.g., cumene) and other materials added with CHP to the reactor. During cumene oxidation small amounts of dimethyl phenyl carbinol (DMPC) and acetophenone are also formed. In the presence of acid catalyst, DMPC dehydrates to alpha-methylstyrene (AMS), a useful by-product. Very high yields of AMS can be obtained

from pure DMPC, e.g., 98 percent yield upon dehydration over acidic silica at 300°C. In the presence of phenol, however, and more specifically in phenol/acetone/cumene which is solvent in decomposition of technical CHP/DMPC mixtures, the AMS yield is normally about 50-60 mol percent of the DMPC. Main by-products are AMS dimers and cumylphenol which have no commercial value. Formation of cumylphenol also reduces the phenol yield.

The present invention includes a process for decomposing a cumene oxidation product mixture containing cumene hydroperoxide (CHP) and dimethylphenyl carbinol (DMPC) to produce phenol, acetone and alpha-methyl styrene (AMS) with enhanced safety of operation and reduced by-product formation which comprises the steps:

(a) mixing the cumene oxidation product in a stirred or back-mixed reactor 1 with an acid catalyst, with 10 to 100 percent acetone relative to the amount of acetone produced during the reaction and with up to 4 weight percent water relative to the reaction mixture, at an average temperature between about 50°C and about 90°C for a time sufficient to lower the average CHP concentration of the reactor to between about 0.2 and about 3.0 weight percent and wherein a portion of DMPC is converted to dicumyl peroxide (DCP); (see Specification page 8 line 33 - page 9 line 15) then

(b) reacting the reaction mixture from step (a) at a temperature between about 120 and 150°C under plug-flow conditions (figure 1, in heater 8 and pipe 9) for a time sufficient to decompose substantially all residual CHP and at least 90% of DCP formed in step (a). See Specification page 9 line 16 - page 10 line 2.

In a preferred process (claim 3 step C) the product from step (b) is submitted to adiabatic flash evaporation (figure 1 evaporator 10), recovering an acetone-rich distillate which is recycled to step (a) to provide said acetone. See Specification page 10 lines 2-10.

It may be preferred that step (a) additionally comprises reacting the reaction mixture having an average CHP concentration of between about 0.2 and about 3.0 weight percent at between 50°C and about 90°C under plug-flow conditions for a time sufficient to produce a reaction mixture having a CHP concentration no greater than about 0.4 weight percent. See claim 2, Specification page 6 line 5-9, page 9 lines 17-25, figure 1 tube 7.

(4) Issues. Whether claims 1-8 are unpatentable over Sifniades et al. in combination with Barilli et al. and Anderson. First, has a prima facie case of obviousness been established. Second, does the evidence presented by appellants in the specification present unexpected results which overcome any presumed prima facie case.

(5) Grouping of Claims. Claims 1, 3 and 5 each represent an important embodiment of the invention not taught or suggested by the cited references.

(6) Argument. The examiner has failed to present a prima facie case of obviousness. The final rejection is over Sifniades et al. (U.S. 4358618) in view of Barilli et al. (U.K. 1,202,687) and Anderson et al. (U.S. 4207264). It is essential to consider the claimed invention as a whole, and whether in light of the cited references one would consider the claimed invention obvious under 35 U.S.C. 103. Panduit Corp. v. Dennison Mfg. Co., 227 USPQ 337 (CAFC 1985); vacated, 475 U.S. 809, 229 USPQ 478 (1986), aff'd on remand 1 USPQ 2d 1593 (CAFC 1987).

Sifniades et al. teaches the use of a three stage operation but without acetone recycle. Barilli et al. and Anderson teach the use of acetone as a solvent, but in a single stage reaction.

The examiner states "The second step in the process of Sifniades et al. is not necessary when acetone is used in the first step since the secondary references show that the cumene hydroperoxide concentration is reduced sufficiently using the

acetone solvent that the plug-flow reaction is not necessary." However, appellants show that surprisingly this is not the case. With reference to the examples of Table I and to Figure 2, examples 1 through 6 and 12, with acetone recycle of 40 and 60%, when plotted in Figure 2 clearly show a steep increase in AMS yield as residual CHP in the back-mixed reactor is allowed to increase from zero up to about 0.2 weight percent, and there is further a more gradual yield improvement as residual CHP increases up to 3 weight percent.

AMS is formed essentially by dehydration of DMPC which is formed along with CHP during oxidation of cumene. The yield of AMS, besides representing the yield of a valuable by-product, is also a measure of the suppression of high-boiling tars. These are chiefly AMS dimers and cumyl phenol. If allowed to persist to the distillation train, DCP is also a source of tars. Cumyl phenol and DCP engender loss of phenol as well as AMS. Therefore, AMS yield, as defined on page 11, lines 10 to 15 of the application, generally correlates with phenol yield.

The AMS yield in example 12 was 62.6% after a single stage reaction in a stirred reactor (p. 11, line 26). No residual CHP was present in the product, therefore this example represents the outcome of a process with acetone solvent as referred to by the examiner and corresponds to the one stage process with acetone shown in the secondary references. The AMS yield increased to 68.1% upon further reaction at 125°C in a tube reactor. This effect is thought to be due to a small amount of DCP (0.55 wt. percent) that was formed in the stirred reactor and decomposed to produce AMS, phenol and acetone in the tube reactor. This result shows that even when operating with acetone solvent with zero residual CHP, a two stage process is beneficial. Nevertheless, we limit our claims to a two stage process in which the residual CHP after the first stage is at least 0.2 wt. percent because the AMS yield is significantly higher in such a process. This result is

respectfully submitted to provide a showing of unexpected results over the teachings of the cited prior art.

A different perspective of appellants' invention is as follows. The prior art teaches that when acetone is used as solvent in CHP decomposition, higher yields of phenol are obtained. However, the mere presence of acetone does not guarantee high yields; other reaction conditions, such as temperature, level of catalyst and residence time must be fine tuned. This is due to the fact that the synthesis of phenol, acetone and AMS from technical CHP stock is a complex process that encompasses several simultaneous and consecutive reactions. Product yields peak out in a rather narrow range of conditions. If reaction in a back-mixed reactor is allowed to proceed beyond optimal CHP conversion, yields decline. Thus the AMS yield (which appellants have shown to correlate with phenol yield) in example 12 of appellants' disclosure could be made much smaller (e.g. 30%) by using more sulfuric acid and/or increasing the residence time. The resulting residual CHP would still be zero. In other words, if the reaction is carried out to complete CHP decomposition, there is no convenient measure of the severity of reaction. Appellants' invention, by specifying that the effluent from the back-mixed reactor contains at least 0.2 wt. percent CHP, ensures that the yield has not peaked out. The reaction is then completed in a second stage in a plug-flow reactor in which residual DCP provides the criterion of the optimal extent of reaction.

In the final rejection the examiner erroneously asserts that "Sifniades et al. teaches the use of acetone in Examples 1 to 5 and 13 to 17". He also states that "applicants have not presented side-by-side comparisons to show the criticality of the instant acetone concentration range".

The examples cited by the examiner do not pertain to the process of decomposing a cumene oxidation product mixture per se taught by Sifniades et al.

Examples 1 to 5 of Sifniades et al. describe the acid catalyzed decomposition of pure CHP. They serve to demonstrate that such decomposition produces increasingly larger amounts of undesired by-products as the temperature is increased. The context of these examples is clearly indicated in the specification of Sifniades et al., col. 4, lines 52 to 56.

Examples 13 to 17 of Sifniades et al. describe the acid catalyzed decomposition of pure DCP. They serve to demonstrate that the best yields of AMS are obtained at temperature over 100°C, e.g. 120°C to 150°. The context of these examples is indicated in the specification of Sifniades, et al., col. 3, lines 1 to 3.

In both series of examples, the solvent consists essentially of equimolar phenol/acetone containing 15 wt% cumene, 1% water, and a catalytic amount (50 to 100 ppm) of sulfuric acid. This mixture is meant to simulate the solvent present during a typical continuous decomposition of cumene oxidation product in which the main products (i.e. equimolar phenol/acetone) along with cumene, added water and catalyst are the main components of the solvent.

Water and catalyst are the only components of the process for decomposition of cumene oxidation product as taught by Sifniades et al. that are added externally. All other components are either present in the original mixture (e.g. cumene) and/or produced during the reaction (e.g. phenol, acetone, part of water). There is no suggestion for addition of acetone in the process. Acetone is used only in examples that serve to illustrate the course of decomposition of specific species (such as CHP and DCP) and then only in essentially equimolar mixture with phenol and with cumene present to simulate a continuous reaction involving no added acetone.

Regarding the second of the examiner's objections, it is respectfully submitted that examples 1 to 12 of the instant application, when viewed according to the plots of Figure 2,

provide a clear side-by-side comparison of the effect of recycling acetone.

There are two objectives in the operation of a commercial process for decomposition of cumene oxidation product that are addressed by the process of the instant application.

- a) High yield of useful products. AMS is a useful product. Moreover, AMS yield as defined on page 11, lines 10 to 15 of the application, generally correlates with phenol yield.
- b) Process stability. The main threat to process stability is a runaway reaction resulting from uncontrolled decomposition of residual CHP as discussed on page 2 lines 36 to 38 and page 3, lines 1 to 11 of the application.

Consequently, it is desirable to operate the decomposition of cumene oxidation product in such a manner that the highest yield is obtained with the lowest amount of residual CHP present. It can be seen from Figure 2 that for a given yield of AMS a much lower concentration of residual CHP is necessary when acetone is recycled in the process than when it is not. Thus, to obtain 80% AMS yield it is necessary to have only ca 0.3 wt% residual CHP when 60% of acetone is recycled, whereas ca 1.7 wt% is required with no acetone recycle. Conversely, for a given level of residual CHP (which corresponds to an accepted level of reactor instability) a higher AMS yield is obtained when acetone is recycled to the process than when it is not. Thus, if residual CHP is set at ca 1 wt%, the AMS yield is ca 84% when 60% of acetone is recycled, whereas it is ca 76% when no acetone is recycled.

The examiner has failed to address the combination including the step(c) expressed in claims 3 and 5. This is not taught or suggested by the cited references.

Serial No. 297,133, Filed January 17, 1989

In view of the discussion above, it is respectfully submitted that the claimed process is not taught or suggested by the cited references, or alternatively that the increase in AMS yield provided by utilization of the second stage provides a showing of unexpected results. Accordingly, it is requested that the Examiner's rejection of claims 1-8 under 35 USC 103 not be sustained.

Respectfully submitted,
RICHARD R. HERTZOG ET AL.

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William H. Thrower
NAME OF APPLICANT, ASSIGNEE, OR APPLICANT'S ATTORNEY
William H. Thrower
SIGNATURE
July 20, 1990
DATE

APPENDIX A (CLAIMS)

WHAT IS CLAIMED:

1. A process for decomposing a cumene oxidation product mixture containing cumene hydroperoxide (CHP) and dimethylphenyl carbinol (DMPC) to produce phenol, acetone and alpha-methyl styrene (AMS) with enhanced safety of operation and reduced by-product formation which comprises the steps:

(a) mixing the cumene oxidation product in a stirred or back-mixed reactor with an acid catalyst, with 10 to 100 percent acetone relative to the amount of acetone produced during the decomposition reaction, and with up to 4 weight percent additional amounts of water relative to the reaction mixture, at an average temperature between about 50°C and about 90°C for a time sufficient to lower the average CHP concentration of the reactor to between about 0.2 and about 3.0 weight percent, and wherein a portion of DMPC is converted to dicumyl peroxide (DCP); then

(b) reacting the reaction mixture from step (a) at a temperature between about 120°C and 150°C under plug-flow conditions for a time sufficient to decompose substantially all residual CHP and at least 90 percent of the DCP formed in step (a).

2. The process of claim 1 wherein step (a) additionally comprises reacting the reaction mixture having an average CHP concentration of between about 0.2 and about 3.0 weight percent at between 50°C and about 90°C under plug-flow conditions for a time sufficient to produce a reaction mixture having a CHP concentration no greater than about 0.4 weight percent.

3. The process of claim 2 further comprising the step:

(c) submitting the reaction product from step (b) to adiabatic flash evaporation to recover an acetone-rich distillate and recycling said distillate to step (a) to provide said acetone.

4. The process of claim 3 wherein the CHP concentration is monitored by on-line analysis.

5. The process of claim 1 further comprising the step:

5 (c) submitting the reaction product from step
- (b) to adiabatic flash evaporation to recover an
acetone-rich distillate and recycling said distillate to
- step (a) to provide said acetone.

10 6. The process of claim 5 wherein the CHP
concentration is monitored by on-line analysis.

7. The process of claim 1 wherein the CHP concentration is monitored by on-line analysis.

8. The process of claim 2 wherein the CHP concentration is monitored by on-line analysis.